

Appendix E

Trend Analysis Case Studies

Introduction

In the following, temporal trends in sediment contamination are discussed by using three case studies reported in literature. These studies include Lake Pepin (downstream of Minneapolis, Minnesota on the Mississippi River), four lakes in the Chattahoochee and Flint River Basin (Georgia), and Puget Sound (Washington State).

Case Studies

Hg Loading to Lake Pepin from the Upper Mississippi River

Introduction

Lake Pepin is a natural lake located on the Mississippi River, 80 km downstream of Minneapolis and St. Paul, Minnesota. Upstream of Lake Pepin, the St. Croix and Minnesota Rivers join the Mississippi River, creating a catchment of 122,000 km² (Figure E-1). The major land use in the Minnesota River Basin is agriculture. Forests and wetlands make up much of the headwater Mississippi (despite the twin cities) and St. Croix river basins. Because of the diverse nature of the three watersheds—Mississippi, Minnesota, and St. Croix—the flows from these watersheds are different, with the nutrient content, suspended sediment, and Hg concentrations being substantially higher in the Minnesota River than in the headwater Mississippi and St. Croix rivers. Lake Pepin has a typical hydraulic residence time of more than 5 days, sufficient to enable settling of suspended matter. Eighty five to ninety percent of the sediment load to Lake Pepin originates from the Minnesota River Basin and consists of fine silt and clays. The location, the hydraulic residence time, and the waters that flow into Lake Pepin make it a repository for sediment and the attached heavy metals. The Mississippi River and Lake Pepin received significant inputs of Hg during the 1960s when the other lakes in the region received no point source loads (Balogh et al., 1997). Balogh et al. (1999) collected sediment cores from Lake Pepin to reconstruct the history of Hg loading in the Upper Mississippi River over the past 200 years. Sediment cores were dated and stratigraphically correlated by using ²¹⁰Pb, ¹³⁷Cs, ¹⁴C, magnetic susceptibility, pollen analysis, and loss-on-ignition. The authors have reported that the Hg stratigraphy in Lake Pepin was undisturbed by post-depositional processes.



Figure E-1. Lake Pepin Location Map.
Source: Balogh et al., 1999.

Total Hg Concentrations in Lake Pepin

Balogh et al. (1999) reported that Hg concentrations in deep presettlement core samples in Lake Pepin varied between 33 and 40 ng/g. These levels of Hg are similar to other lakes in the region and are comparable to the surface soil Hg concentrations of 18 to 44 ng/g in the Minnesota River basin. Results from sediment cores indicate increased Hg concentrations after 1800, when the European settlers established their continuous presence along the Minnesota and Mississippi rivers. By 1900, the Hg concentration in sediments was above 200 ng/g, and most cores indicated a steady or decreasing Hg concentration between 1920 and 1950.

However, between 1949 and 1968, Hg concentrations began to increase, reaching peak values of Hg ranging from 452 to 669 ng/g. The concentration of Hg in sediment cores declined substantially from the late 1960s and continued to decrease to concentrations between 106 to 161 ng/g as presented in Table E-1.

Table E-1. Total Hg Concentrations (ng/g) in Lake Pepin Sediment Cores.

Core Number	Presettlement	Peak	Modern
1	33	662	106
2	34	669	120
3	34	528	120
4	33	561	128
5	40	519	121
6	36	503	135
7	38	570	123
8	34	499	128
9	37	452	137
10	39	492	161

Source: Balogh et al., 1999.

Averaging the sediment core data by 10-year intervals, Balogh et al. (1999) estimated a loading rate of 3 kg/yr to characterize naturally occurring deposition of Hg under pristine conditions before European settlement began ca. 1830. Hg deposition progressively increased during the 19th and 20th centuries, with about one-half of the total Hg load deposited from 1940 to 1970 and the peak accumulation rate of 357 kg/yr identified during the 1960s.

As a result of decreasing the discharges of Hg from municipal and industrial wastewater plants, the Hg deposition rate in Lake Pepin has declined by almost 70 percent from the maximum loading during the 1960s to 110 kg/yr during 1990-1996 as seen in Figure E-2. However, the current Hg loading rate of 110 kg/yr for Lake Pepin is substantially higher than the estimated presettlement loading rate of 3 kg/yr.

The average Hg loading rate in the Mississippi River immediately upstream of Lake Pepin was reported as 102 kg/yr by Balogh et al. (1999). This indicates that much of the mercury loading from the Upper Mississippi River is deposited with the sediment into Lake Pepin.

Summary and Conclusions

The accumulation of Hg to Lake Pepin can be attributed to urban runoff and industrial/domestic users. In addition to the Hg loading due to atmospheric deposition from coal combustion and industrial emissions, Balogh et al. (1999) have identified other possible sources of Hg which might have contributed to the

**Historical Mercury Loading
Lake Pepin Sediments**

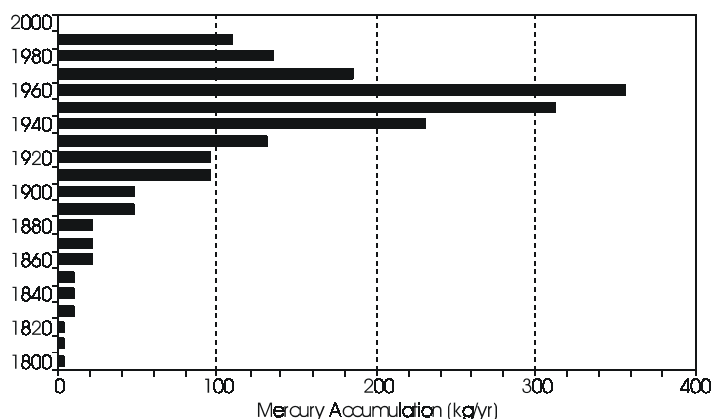


Figure E-2. Historical Mercury Loading Rates in the Upper Mississippi River Reconstructed from Sediments of Lake Pepin. Source: Balogh et al., 1999.

accumulation in Lake Pepin. During the period of initial European settlement, Hg was used extensively for medicinal purposes, in photography, in dentistry, in tanning/dyeing industries, and in thermometers. In the 1900s the medicinal use of Hg decreased but its industrial use continued to escalate.

Balogh et al. (1999) have suggested that the inability of Metropolitan Wastewater Treatment Plant, located along the Mississippi River at St. Paul, Minnesota, to cope with the increased population might have contributed to the increased Hg loading. The capacity of the treatment plant was increased and a secondary treatment unit was added during 1966-1968. The enhanced treatment capacity and the diminished use of Hg after the 1960s are reflected in the substantial decline of Hg loadings to Lake Pepin, as shown in Figure E-2. Although the investment in water pollution control has been very successful in reducing Hg in the Upper Mississippi River, ambient levels are still 30 times greater than the pristine conditions of the early 1800s (MCES, 2000).

Historical Trends in Organochlorine Compounds from Four Georgia Lakes

Introduction

One of the major objectives of the National Water-Quality Assessment Program (NAWQA) of the U.S. Geological Survey (USGS) is to study and define historical trends of water quality in the waters of United States. As a part of the NAWQA program, Van Metre et al. (1997) studied temporal trends of PCBs, total DDT, and chlordane concentrations using sediment cores in four lakes in Georgia (Figure E-3). The study used radiochemical dating of sediment cores and measurement of chlorinated organic compounds by standard extraction techniques. Other results for a lake in Texas and a reservoir in Iowa can be found in Van Metre et al. (1997).

Lake Harding is located along the Chattahoochee River 184 km downstream of Atlanta and receives drainage from metropolitan Atlanta. The Chattahoochee River flows from northeast Georgia through metropolitan Atlanta and is a major water supply source and receptacle for wastewater disposal for Atlanta. The drainage to Lake Harding was reduced when West Point Lake, 25 km upstream of Lake Harding, was constructed in 1974. Lake Walter F. George is located on the Chattahoochee River, 290 miles downstream of Atlanta and is separated from metropolitan Atlanta by Lake Harding and West Point Lake. Lake Blackshear is located 314 km downstream from Atlanta along the Flint River. Land use in the drainage area for Lake Blackshear is agriculture and forestry. Lake Seminole is located at the confluence of Flint and Chattahoochee Rivers, forming the Apalachicola River. The drainage area for Lake Seminole is mainly agricultural.

In all four locations, i.e., Lakes Harding, Walter F. George, Blackshear and Seminole, Van Metre et al. (1997) reported uniform, fine-textured sediments with no evidence of bioturbation, i.e., no major displacements within the unconsolidated sediments by benthic organisms. In addition, the authors have reported no post-depositional mixing of the sediment by current or waves in the

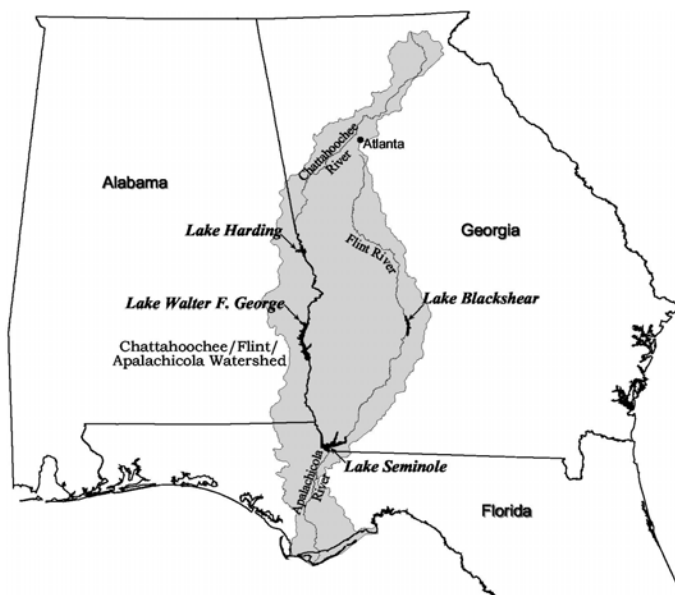


Figure E-3. Location Map of Upper Chattahoochee Basin.

lakes. This conclusion was feasible by virtue of pronounced ^{137}Cs peaks with exponential decreases in ^{137}Cs to the sediment surfaces at the lakes.

Trends in PCBs

Results of the findings by Van Metre et al. (1997) for sediment concentrations of PCBs are presented in Table E-2. The peak PCB concentration during 1950 to 1969 in Lakes Harding and Walter F. George near Atlanta, Georgia are higher than in Lakes Blackshear and Seminole in rural Georgia. Lakes Harding and Walter F. George receive effluent from metropolitan Atlanta, whereas the drainage area for Lakes Blackshear and Seminole are mainly agricultural and forested areas.

Table E-2. PCB Concentrations ($\mu\text{g}/\text{kg}$) in Lake Sediments.

Period/ Location (State)	Lake Harding	Walter F. George	Lake Blackshear	Lake Seminole
	Georgia	Georgia	Georgia	Georgia
Before 1950	~50	NA	NA	NA
1950-1969	280-380 ^a	220 ^b	8	3-10
1990s	42-46	<14	1	10
% Decrease	85-88	94	88	(70)-0

^a between 1950-1966

^b in 1968

Source: Data extracted from Van Metre et al., 1997.

Drainage and effluent from metropolitan Atlanta was directed to Lake Harding until 1974, when West Point Lake, upstream of Lake Harding, was constructed to intercept a portion of the flow. Van Metre et al. (1997) have reported that PCB concentrations in seven sediment core samples at West Point Lake ranged from 110 $\mu\text{g}/\text{kg}$ to 32 $\mu\text{g}/\text{kg}$ for the period - 1974 to the 1990s. The concentrations of PCB during the corresponding time period at Lake Harding are similar (Table E-2). This led them to conclude that the substantial decrease in PCB concentrations in sediments in Lake Harding between the 1970s and the 1990s is not due to West Point Lake intercepting a significant PCB load from upstream. The decreases are due to reduced manufacture and uses of PCB, and its regulation in the United States. The authors have attributed the spatial trend of PCB concentrations in the sediments of Lake Seminole partly to the location of the coring site.

Trends in Total DDT

Sediment concentrations of DDT and its derivatives (DDD and DDE) found in core samples in the four lakes are shown in Table E-3. Like the temporal variation of PCBs in the lake sediments, total DDT concentrations showed steep decline in all lake sediments presented here. This decline is consistent with the use of DDT in the United States, with peak usage in the early 1960s until the use of DDT was prohibited nationally in 1973. DDT concentrations in West Point Lake sediments follow a similar pattern to the two downstream lakes, Lakes Harding and Walter F. George, after early 1970s. As in the case of PCBs, the decline in DDT concentrations in Lake Harding is not due to West Point Lake intercepting a portion of the DDT load, before it reaches Lake Harding. Actual reduction in usage and the banning of DDT, resulted in reduced concentrations of total DDT in lake sediments.

Table E-3. DDT Concentrations (µg/kg) in Lake Sediments.

Period/ Location	Lake Harding	Walter F. George	Lake Blackshear	Lake Seminole
	Georgia	Georgia	Georgia	Georgia
1940s-1965	35	74 ^a	57	30
1990s	5.9	5.5	6.5	7.5
% Decrease	83	93	89	75

^a in 1968

Source: Data extracted from Van Metre et al., 1997.

Trends in Chlordane

Used as a pesticide in agricultural and residential areas, chlordane was introduced for agricultural use in the United States in the early 1970s and was used extensively until 1974, when its agricultural use was banned. However, it continued to be used in termite control until 1988 (Van Metre and Callender, 1997). The only permitted uses of chlordane after 1988 were in power transformers, for fire ant control and the use of existing stocks by homeowners. Sediment concentrations of chlordane from the study by Van Metre et al. (1997) are presented in Table E-4. Similar to PCBs, chlordane concentrations are higher in urban lakes by virtue of their use in such surroundings. Unlike PCBs and total DDT, reaching peak values during the 1960s, chlordane concentrations peaked during the late-1980s or the 1990s. Chlordane showed a minor decline during sampling in 1994/95. This may be due to the more stringent control of the use of chlordane even in urban areas, as permitted under current regulations.

Van Metre et al. (1997) have reported that chlordane concentrations in West Point Lake ranged between 20 to 55 µg/kg. These concentrations are higher than the chlordane concentration in Lake Harding prior to 1974. Hence the substantial decrease in chlordane concentration in Lake Harding is due to West Point Lake serving as a sink for chlordane from urban Atlanta and other enhanced sources of chlordane.

Table E-4. Chlordane Concentrations (µg/kg) in Lake Sediments.

Period/ Location	Lake Harding	Walter F. George	Lake Blackshear	Lake Seminole
1960s	46-84 ^a	6 ^c	ND	ND
1990s	6-8 ^b	NA	ND	ND
% Decrease	87-90	NA	NA	NA

^a 1950-1970.

^b Early 1970s.

^c In 1973.

ND: not detected.

NA: not available.

Source: Data extracted from Van Metre et al., 1997.

Summary and Conclusions

The temporal decline of PCB, and ΣDDT in the lake sediments are parallel to the usage and regulation of these chemicals in the United States. Eisenreich et al. (1989) have reported similar results on sediment cores from Rochester Basin of eastern Lake Ontario. Steep increases in PCB concentration in sediments occurred between 1940s and 1960s with peak values between 1966-1969. These concentrations decreased substantially by 1980. Sanders et al. (1992) studied sediment cores from a lake

in rural England. Their study concluded similar declines in PCB and total DDT concentrations from peak values during the respective peak production and usage periods. However, Eisenreich et al. (1989) and Sanders et al. (1992) have alluded to the possibility of sediment mixing by various processes and thus distorting the historical records of contaminant deposition. In addition, Sanders et al. (1992) have reported that PCB and DDT in sediment cores were recorded, prior to their use in the United Kingdom. The authors have suggested long-range atmospheric transport from the United States, and mainland Europe as possible sources for such contamination.

Accumulation of Chemicals in Puget Sound

Introduction

Puget Sound, an estuary located in northwestern Washington State (Figure E-4), was one of the first areas in the United States to be studied extensively for sediment contamination. Early studies from the 1980s demonstrated fairly extensive sediment contamination, especially near major industrial embayments (Dexter et al., 1981; Long, 1982; Malins et al., 1980; Riley et al., 1981). These early assessments demonstrated that Puget Sound sediments were contaminated by many organic and inorganic chemicals, including PCBs, PAHs, and metals. Although contaminant concentrations in sediments tended to decrease rapidly with distance from the nearshore sources, researchers also documented widespread low-level contamination in the deepwater sediments of the main basin of Puget Sound (Ginn and Pastorok, 1982). Also in the 1980s, several kinds of biological effects, including cancerous tumors, were reported in organisms from contaminated areas of Puget Sound (Becker et al., 1987).

Although contaminant levels in some surface sediments have started to decrease since pollution controls were established in the last few decades, contamination in the deep central Puget Sound basin are still significantly higher than estimated preindustrial levels. In urban areas, present levels of contaminants are up to 100 times the levels in the cleanest rural bays. Sediment samples collected from many locations in Puget Sound, such as Bellingham Bay, Commencement Bay, Port Gardner Bay, Elliot Bay and Eagle Harbor, were observed to be toxic to test organisms. In 1985 the Washington State legislature established the Puget Sound Water Quality Authority (PSWQA). The PSWQA has exercised greater control of point source discharges to the sound by mandating industries to reduce the chemical concentrations of their discharges.

Scientists from the Battelle/Marine Sciences Laboratory (MSL) in Sequim, Washington collected sediment cores from six locations in the main basin of Puget Sound during 1991. The location of these sites was based on minimal disturbances reported due to natural or anthropogenic activity in these areas. The study analyzed PAHs, PCBs, metals, and pesticides using sediment cores. The field sampling techniques, the different analytical methods and the data analysis used for the various classes of chemicals listed above are described in the National Oceanic and Atmospheric Administration Technical Memorandum NOS ORCA 111 (Lefkovitz et al., 1997). Recovery rates for the various contaminants were computed based on the decline from a peak concentration to the background level over a time period. Sediment ages were calculated by ignoring the sediment density. The study used both ^{210}Pb and



Figure E-4. Location map of Puget Sound.

^{137}Cs dating. By virtue of a distinct subsurface maximum of ^{137}Cs , the authors have ruled out the possibility of sediment disturbance by mixing and migration. A trend analysis was also carried out for metals, PCBs and DDT at three core locations to evaluate the statistical significance of the relationship between the concentrations of the various contaminants and the time period from 1970 to 1991. A decrease in concentration of the contaminants during this 20 year period—at a predetermined confidence level (95%, i.e., $\alpha=0.05$)—served as empirical evidence of the effectiveness of the environmental legislation enacted during this period.

Of the 16 metals analyzed, eight showed reproducible temporal trends of increasing concentrations to a peak followed by a decline through the present time. The peak and surface (background) concentrations of metals showing a temporal trend from three locations and the year associated with these concentrations are listed in Table E-5. The concentrations listed in Table E-5 were extracted from profiles of concentration versus year at the various core sites.

Table E-5. Maximum and Surface Concentrations ($\mu\text{g/g}$) of Selected Metals for Three Core Locations Collected During 1991.

Metal	Ag	As	Cu	Hg	Pb	Sb	Sn	Zn
Location 1								
Max Year	1965	1965	1960	1949	1965	1960	1965	1965
Max	0.91	19.5	54.6	0.479	48.9	2.05	4.9	134.6
Surface	0.68	12.5	42.7	0.179	30.3	1.28	3.94	114.7
% Decrease	25.3	35.9	21.8	62.6	38.0	37.6	19.6	14.8
Recovery Rate	0.009	0.280	0.397	0.007	0.744	0.026	0.038	0.796
Location 2								
Max Year	1982	1964	1947	1947	1922	1952	1962	1962
Max	0.84	28.3	70	0.505	69.4	3.9	4.85	167.7
Surface	0.69	13.1	49.3	0.213	36.7	1.6	3.96	119.2
% Decrease	17.9	53.7	29.6	57.8	47.1	59	18.4	28.9
Recovery Rate	0.017	0.563	0.470	0.007	0.474	0.059	0.031	1.672
Location 3								
Max Year	1965	1950	1963	1950	1954	1963	1963	1954
Max	0.65	23.5	64.7	0.403	62.3	2.43	4.25	128.8
Surface	0.59	17.3	52.7	0.28	44.7	1.52	2.78	115.5
% Decrease	9.2	26.4	18.5	31.3	28.3	37.4	34.6	10.3
Recovery Rate	0.002	0.155	0.444	0.003	0.489	0.034	0.054	0.369
Average Recovery Rate	0.009	0.333	0.437	0.006	0.569	0.039	0.041	0.946
95%CL \pm	0.014	0.424	0.076	0.005	0.308	0.035	0.025	1.348

Source: Lefkovitz et al., 1997.

The concentration of lead in all sites peaked during the period 1920 to 1960, followed by a declining trend from 1960 to the present. In addition to the recovery rate, lead also showed decreasing trend with time for the period from 1970 to 1991. The authors of this study have attributed the effectiveness of the current environmental regulation for this declining trend. The concentration of arsenic peaked between the early 1950s and the 1960s. The maximum concentration of arsenic was found on two locations close to a copper smelter operated from 1889 to the 1980s in Tacoma, Washington. Crecelius et al. (1975) reported the smelter as the major anthropogenic source of arsenic and antimony, both by-products of the smelting operation. Concentrations of antimony showed an increasing trend between 1900 and the 1950s followed by a decline in concentration to the time of sampling. Though both metals showed percentage

decreases greater than 25 percent, only antimony had a statistically significant recovery rate of 0.039 at the 95 percent confidence level. Moreover, antimony had a decreasing trend with time, whereas concentrations of arsenic remained unchanged during this period. Levels of Hg in all three locations decreased by an average of 50 percent from a peak concentrations during late 1940s, and mercury had a sediment recovery of 0.006 at 95 percent confidence level. However, from the trend analysis for the period from 1970 to 1991, the level of Hg remained the same with no statistically significant decrease with time.

Silver showed an average recovery of 17 percent from peak values in the three locations. The trend analysis did not show a significant correlation between time period and concentration for silver except at location 3. Copper and zinc produced a statistically significantly decreasing trend with time. The sediment recovery rate for Cu was significant at 95 percent confidence level whereas Zn did not show a significant recovery rate. Tin showed an average decrease of 24 percent in all three locations and an average recovery rate of 0.041 ± 0.025 $\mu\text{g/g}$ per annum. However, concentrations have not shown a declining trend since wide fluctuations are still reflected in surface sediments. For most metals the highest reductions in concentrations were detected at location 2, which is in the vicinity of many sewage treatment plant outfalls. Because secondary treatment of sewage discharge to Puget Sound was initiated after the late 1950s, the decrease in metal concentrations can be attributed to these treatment techniques.

In addition to metal concentrations in sediment cores, the study included concentrations of PAHs, PCB, and DDT. Details of the levels of the various contaminants are shown in Table E-6. Concentrations of total PAH varied from 100 ± 21.6 $\mu\text{g/g}$ at the lower levels of the sediment to a maximum of 6,788 $\mu\text{g/g}$ during the early 1940s, subsequently declining to an average of 1,300 $\mu\text{g/g}$ at the time of sampling in 1991. Neither the sediment recovery rate nor the correlation between time period and the concentration of total PAHs were statistically significant at the preselected 95 percent confidence level. The temporal trend in PAHs in the sediment is consistent with the use of fossil fuel in two cities, Seattle and Tacoma in Washington State during the time period. Barrick (1982) has reported that combined sewage and stormwater effluent and atmospheric deposition are the major sources of PAH contamination of the sediments of Puget Sound. Combustible PAHs derived from combustion of fossil fuels and other organic-rich materials such as wood have a high molecular weight and consist of four to six aromatic ring compounds. Like the total PAHs, the combustible PAHs had neither the sediment recovery rate nor the decreasing trend between time period and concentration.

Both PCBs and DDT showed trends similar to those of metals and PAHs. The PCB concentration in Puget Sound sediments appeared from the 1930s to peak values until the mid-1970s when the use of PCBs was restricted in the United States. In spite of an average reduction of 68 percent from peak values of PCB in all three locations, in only one location was a statistically significant decrease with time was recorded for sediments deposited after 1970 to the present. Moreover, the estimated sediment recovery rate of $0.649 (\pm 0.653)$ $\mu\text{g/g}$ was not statistically significant at the 95 percent confidence level. DDT concentrations were reduced by an average of 42 percent in the three locations. In only one location there was a decreasing trend in DDT concentrations for sediments deposited after 1970. The average recovery rate for DDT in all three core locations was $0.103 (\pm 0.096)$ $\mu\text{g/g}$ per year.

Summary and Conclusions

The concentrations of Cu, Pb, Sb, and Zn declined in Puget Sound at statistically significant levels from 1970 to the time of sampling in 1991. Ag, Hg, As and Sn did not exhibit a similar declining trend in sediment contamination. Moreover, the average recovery rates (i.e., decrease in sediment contamination levels) for As, Ag, and Zn was not significant between the peak period and time of sampling. Among the organic chemicals studied, only DDT showed a statistically significant recovery rate, whereas the recovery rates for PCBs, total PAH and combustible PAH had wider margins of variation. The authors of this study have not given significance to the recovery rates reported for Hg, since the analytical error

Table E-6. Maximum and Surface Concentrations (µg/g) of Selected Organic Contaminants for Three Core Locations Collected During 1991.

Organic Contaminant	PCB	DDT	Total PAH	Combustible PAH
Location 1				
Max Year	1960	1960	1943	1943
Max	34.5	4.71	6788	5917
Surface	9.00	1.19	1434	1162
% Decrease	73.9	74.8	78.9	80.4
Recovery Rate	0.851	0.117	114	101
Location 2				
Max Year	1966	1984	1942	1942
Max	25.8	5.76	3430	2898
Surface	5.33	4.77	1303	1050
% Decrease	79.3	17.2	62	63.8
Recovery Rate	0.819	0.142	43.412	37.878
Location 3				
Max Year	1961	1961	1935	1935
Max	15.5	4.25	1883	1516
Surface	7.39	2.8	1212	977
% Decrease	52.2	34.2	35.6	35.5
Recovery Rate	0.278	0.050	12.198	9.793
Average Recovery Rate	0.649	0.103	56.5	49.6
95% CL ±	0.653	0.096	106	95

Source: Lefkovitz et al., 1997.

could be greater than the actual concentrations of Hg observed in this study. Though the declining trend in sediment contamination for certain chemicals is an encouraging sign, it is essential to note that there are some areas of concern with certain classes of chemicals. These may be attributed to either previous use and disposal practices or due to chemicals discharged currently to Puget Sound from direct or indirect sources. Puget Sound is surrounded by Seattle and Tacoma, two fast developing and heavily industrialized regions. Hence it is essential to control future discharges to the area, as well as to implement the best available technology, and monitor the water quality.

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